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19. ABSTRACT (Continue on reverse if necessary and identify by block number) The first operational semiconductor diode lasers were demonstrated in the summer of 1991 independently by two US groups, one at 3M and the other a team effort shared by Purdue and Brown Universities. As a result of the close collaboration between MBE and TEM groups within this grant, the structures for lasing and LED (as well as display device) operation were realized with the lowest defect concentrations ever reported for II-VI structures grown on GaAs by MBE. The reduction of the dislocation levels resulted from an iterative process where the growth could be modified in response to the TEM analysis. The AFOSR funded interface studies have led to our appreciation of the electrical and microstructural considerations obtaining at II-VI/III-V heterovalent interfaces. As a result, the Purdue/Brown group has had equal success in making laser diodes with substrates of both doping types. The Purdue/Brown collaboration has obtained CW operation at 77K as well as pulsed operation at room temperature using a Zn(S,Se)-based device configuration emitting in the blue (490nm at room temperature). Laser power output per facet for such devices can exceed 300mW in a pulsed mode, and threshold current densities are below 200A/cm². In addition to diode lasers, efficient LED devices as well as flat panel display devices were fabricated. The blue LEDs provide power outputs in excess of 100μW while exhibiting external quantum efficiencies of 0.1% at room temperature. We have found that the approach used here for the p-doping of Zn(S,Se) by nitrogen, can also be used to obtain free hole concentrations approaching 10¹⁹ cm⁻³ in ZnTe. We have designed and grown a multilayer structure, consisting of 17 cells, each of 20 Å thickness, to implement a graded contact structure. The proposed contacting scheme was evaluated by means of a transmission-line model measurement of specific contact resistance. The specific contact resistance of the graded contact was in the range (10⁻⁴-10⁻³ Ω·cm²) which is usually considered acceptable for large contact area devices such as LEDs and laser diodes. The graded Zn(Te,Se) graded contact structure was employed in Zn(S,Se)-based LED and laser device structures. The laser diode forward bias voltage for lasing was reduced to 15-17V (from ~30V). These lasers have been thus far operated at temperatures in excess of 250K.

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"II-VI SEMICONDUCTOR SUPERLATTICES"

by

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The past two years in many ways have been the most important period in the 30 year long research effort to develop the II-VI semiconductors for application to photonic devices. Despite significantly larger efforts world wide than those in the US, the first operational semiconductor diode lasers were demonstrated in the summer of 1991 independently by two US groups, one at 3M and the other a team effort shared by Purdue and Brown Universities. The Purdue project has been funded partially by AFOSR. AFOSR has especially promoted the close collaboration of the MBE and TEM groups at Purdue. As a result of this close collaboration, the structures for lasing and LED (as well as display device) operation were realized with the lowest defect concentrations ever reported for II-VI structures grown on GaAs. The unprecedented reduction of the dislocation levels achieved in the optical structures resulted from an iterative process whereby the results of a specific growth scheme was evaluated by TEM (often within 2 or 3 days after growth), such that the growth could be modified (by steps discussed by both MBE and TEM participants) in response to the TEM analysis. Further, the AFOSR funded interface studies have led to our appreciation of the electrical and microstructural considerations obtaining at II-VI/ III-V heterovalent interfaces. It is significant that while 3M has been unable to utilize structures grown on p-GaAs substrates where holes are injected into the ZnSe-based layers from the substrate; the Purdue/Brown group has had equal success with substrates of both doping types.

The details of the overall grant research activity for the first two years are described in the previous Annual Reports for this grant, and in the numerous publications listed in this final report. In addition, some key publications/preprints of the past year are appended. In sections A and B below we have presented some highlights of the past year, while part C includes a three-year overview of the specific effort involving transmission electron microscopy.

A. Recent device developments involving blue laser diodes and LEDs

Since the reporting of the first prototype devices, the field has moved rapidly in the further development of both laser diodes and LED display devices. In particular, CW operation has been obtained at 77K as well as pulsed operation at room temperature using a Zn(S,Se)-based device configuration emitting in the blue (490 nm at room temperature). Laser power output per facet for such devices can exceed 300 mW in a pulsed mode, and threshold current densities are below 200A/cm^2 . The Brown/Purdue laser device data is for laser diodes employing charge injection from both n and p-GaAs substrates. In addition to diode lasers, efficient LED devices as well as flat panel display devices were fabricated. The blue LEDs provide power outputs in excess of $100\mu\text{W}$ while exhibiting external quantum efficiencies of 0.1% at room temperature. By employing coated facets, the room temperature laser diode operation is associated with a current threshold which has been reduced from our previously reported 1600A/cm^2 to a value of 1000A/cm^2 , which is quite a respectable number, and one similar to III-V laser diode performance. The challenge is to achieve room temperature operation in the CW mode. The problem has been

device failure due to excessive heating, primarily arising from poor contacts to p-ZnSe. One of our most important contributions of the grant period relates to the issue of ohmic contacts to p-ZnSe and is discussed below.

B. The first ohmic contacts to p-ZnSe.

The difficulty in fabrication of low resistance ohmic contacts to the p-ZnSe layers has been a potential obstacle to the technological success of LEDs and diode lasers. At present, the usual method for contacting p-ZnSe is the deposition of gold to form a Schottky contact having a relatively low barrier.

We have recently reported that the approach used here for the p-doping of Zn(S,Se) by nitrogen, can also be used to obtain the most effective p-doping yet reported for the MBE growth of ZnTe [9]. Free hole concentrations approaching 10^{19}cm^{-3} were obtained. At the same time good crystalline quality, as revealed by x-ray rocking curves and photoluminescence measurements, was maintained. The observation that gold forms a low resistance contact to nitrogen doped ZnTe epilayers, suggests the use of heavily p-doped ZnTe as an intermediate layer for contacting p-ZnSe. However, if one examines the expected energy band lineup at a p-ZnTe/p-ZnSe interface, it is seen that the valence band offset ($\sim 1\text{eV}$) between ZnTe and ZnSe forms a barrier to hole injection in the form of a valence band "spike". A means for removing the energy spike in the valence band is to introduce a Zn(Se,Te) layer having a graded bandgap. We have designed and grown a multilayer structure, consisting of 17 cells, each of 20 Å thickness, to implement such a graded structure. In each cell both the thicknesses of ZnTe and ZnSe layers are varied to approximate a graded region. The first cell next to the p-ZnSe epilayer contained 18 Å of p-ZnSe and 2 Å of p-ZnTe [10], the next cell 17 Å of p-ZnSe and 3 Å of p-ZnTe, and so on. The proposed contacting scheme was evaluated by means of I-V characterization, a transmission-line model (TLM) measurement of specific contact resistance, and by employing the graded contact in actual laser diodes and LED devices. The specific contact resistance of the Au/p-ZnTe/graded layer/p-ZnSe contact was determined by a standard TLM measurement. The specific contact resistance was determined to be approximately $4 \times 10^{-3} \Omega\text{-cm}^2$. (A similar technique was applied to the Au/p-ZnTe contacts yielding a value of about $1 \times 10^{-3} \Omega\text{-cm}^2$.) The specific contact resistance of the graded contact was in the range (10^{-2} - $10^{-5} \Omega\text{-cm}^2$) which is usually considered acceptable for large contact area devices such as LEDs and laser diodes.

In order to demonstrate their robust character, the graded Zn(Te,Se) graded contact structure was employed in Zn(S,Se)-based LED and laser device structures. The LED forward conduction was seen to appear at 3-4 V at 77K. At the threshold of the current turn-on, immediate visual evidence of LED action was observed, and the output level exceeded 100 μW at 4 V. For these devices, the efficiency of the LED was reduced by absorption in the top 2000 Å thick ZnTe layer. We specifically mention the LED performance only at 77K since our earlier work has shown that at this temperature the recombination process is dominated by the radiative recombination [6]. (This

LED, as in the case of our previously reported LEDs, has satisfactory performance at room temperature under dc operation, but now has a much lower operating voltage.) The point is that the turn-on voltage for forward conduction results in a current which actually represents the injection of electron-hole pairs into the QWs for radiative recombination. In the absence of the correspondence between the onset of current flow and the appearance of optical emission, the possibility always exists that an apparently low turn-on voltage could be due to a spurious conduction mechanism which is influenced by defects in the pn-junction devices. For the laser diode with a graded contact, the forward bias voltage for lasing was reduced to 15-17V (from ~30V). These lasers have been thus far operated at temperatures in excess of 250K. The current thrust of the diode laser effort is to achieve room temperature operation under continuous bias, and with long life.

C. Specific Summary of the TEM Program

During the year from 1989 to 1990, the TEM program has been focused on interfacial atomic structures of II-VI/III-V compound heterostructures and related systems. The main results were obtained from the studies of ZnSe/GaAs, CdTe/InSb and Ga₂Se₃/GaAs interfaces.

The change of the atomic structure of the ZnSe/GaAs interface with the surface stoichiometry of GaAs was suggested by our C-V measurements of interface state densities of the MIS structures. The first direct evidence for such a structure was obtained by TEM observations of a series of ZnSe/GaAs interfaces. For the study, three different types of ZnSe/GaAs epilayer-epilayer heterostructures were grown by selecting the surface stoichiometry of GaAs epilayers using a modular MBE system. The first heterostructure was grown on an As-rich surface which exhibited a c(4x4) reconstruction, the second heterostructure grown on a surface whose As coverage was intermediate and exhibited at (4x6) reconstruction, and the third heterostructure grown on an As-deficient surface which exhibited a (4x3) reconstruction. A set of 200 dark field images were taken from cross-sectional samples of the three heterostructures under the identical imaging condition. A clear trend was found from these images where a more distinct bright line appears at the ZnSe/GaAs interface as the GaAs surface becomes increasingly As-deficient. Through the analysis of dark field images taken with different reflections, the bright line was identified as an interfacial compound layer having a structure identical to that of Ga₂Se₃. Ga₂Se₃ is known to crystallize in a zincblende structure with one third of Ga sites being vacancies. Reported thermodynamic data such as the heat of atomization of Ga₂Se₃ show that this compound is a highly stable phase compared to other compounds which may form at the ZnSe/GaAs interface, supporting the results of our TEM study.

We have obtained similar results from the TEM observation of the CdTe/InSb epitaxial interface which is another combination of closely lattice matched II-VI and III-V compounds. Earlier studies by Raman spectroscopy suggested the existence of In₂Te₃ at the CdTe/InSb interface. Similarly to Ga₂Se₃, In₂Te₃ forms a zincblende type structure having vacancies in the In sublattice. Results of the TEM studies of the ZnSe/GaAs and CdTe/InSb systems suggest that

the formation of a III-VI interfacial compound layer may commonly occur at a variety of II-VI/III-V compound systems.

During the course of the research on ZnSe/GaAs interfaces, we have found highly developed vacancy ordering at a Ga₂Se₃/GaAs epitaxial interface. The study of this interfacial vacancy ordering is expected to deepen our understanding of the nature of chemical bonds at II-VI/III-V compound interfaces, as the existence of III₂VI₃ compounds at II-VI/III-V compound interfaces was confirmed in our recent study.

Distinct superstructure reflection rods resulting from the interfacial ordered structure are observed in cross-sectional electron diffraction patterns. These superstructure reflection rods appear as weak spots in plan-view electron diffraction patterns, the locations of which correspond to the c(2x2) superstructure. Direct images of the interfacial ordered structure were obtained in cross-sectional HRTEM images as a periodic arrangement of bright spots along the Ga₂Se₃/GaAs interface with a period of 5.65 Å. A structure model of the interfacial ordered structure was derived by the analysis of diffraction patterns and HRTEM images. In the model, half of the sites of the interfacial Ga plane are left as vacancies, forming a c(2x2) ordered arrangement.

During the years from 1990 to 1992, the main effort of the TEM program was directed to microstructural analyses of ZnSe based LED and laser structures. Numerous structures having different layer configurations have been examined by TEM. The results of these analyses have been used for optimizing the structures and growth conditions in the MBE program. The main finding in the TEM studies of these structures is the importance of the use of closely lattice matched substrates/buffers for the growth of wide-gap II-VI compound structures. This need is compounded by the fact that these compounds are considerably softer than III-V and elemental semiconductors, being highly susceptible to the formation and multiplication of lattice defects. Once lattice defects have formed at an interface due to a lattice mismatch, they easily thread into the II-VI compound layers and are difficult to be eliminated or terminated, compared to those in III-V and elemental semiconductors.

One group of structures studied extensively by TEM are ZnSe/ZnCdSe based LEDs grown on In_xGa_{1-x}As buffer layers which were intended to serve as closely lattice matching substrates. Both cross-sectional and plan-view TEM observations showed that InGa_{1-x}As buffer layers having very low dislocation densities ($10^4 \sim 10^5 \text{ cm}^{-2}$) were obtained by growing on GaAs substrates. Significant degrees of tetragonal distortions of the buffer layer crystals which resulted from the partial relaxation of lattice mismatches were found by the X-ray diffraction and TEM observations. ZnSe/ZnCdSe LED structures having high structural quality with dislocation densities in the lower range of 10^6 cm^{-2} were obtained by using In_{0.045}Ga_{0.955}As buffers the in-plane lattice spacings of which were slightly smaller than that of ZnSe.

In the more recent work, we have examined ZnSe_{1-x}S_x laser structures grown directly on GaAs layers. The sulfur contents in the structures were controlled to be close to that which gives lattice matching with GaAs. TEM images as well as FWHM of X-ray rocking curves showed that

these structures have extremely high structural quality, approaching that of dislocation-free perfect crystals. Only in plan-view samples were we able to find lattice defects, the densities of which were in the range of 10^5 cm^{-2} . To our knowledge, these are the lowest defect densities obtained to date in wide-gap II-VI compound layers. A number of laser structures consisting of $\text{ZnSe}_{1-x}\text{S}_x$ buffer and cap layers and ZnSe active layers have been analyzed. In all those samples, defect densities in $\text{ZnSe}_{1-x}\text{S}_x$ layers were found to be lower than those in the ZnSe layers by one order of magnitude. This observation suggests that the addition of sulfur atoms to a ZnSe crystal increases the hardness of the crystal, and hence, makes the formation of dislocations more difficult than in the binary crystal, similarly to the case of the addition of In atoms to a GaAs crystal, which implies the possibility of a new approach for reduction of defect densities in wide-gap II-VI compound structures.

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APPENDIX

A graded band gap ohmic contact to p-ZnSe

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We describe a low-resistance quasiohmic contact to p-ZnSe which involves the injection of holes from heavily doped ZnTe into ZnSe via a Zn(S_{0.5}Te_{0.5}) pseudograded band gap region. The specific contact resistance is measured to be in the range of $2-8 \times 10^{-3} \Omega \text{ cm}^2$. The graded heterostructure scheme is incorporated as an efficient injector of holes for laser diode and light emitting diode devices, demonstrating the usefulness of this new contact scheme at actual device current densities.

The successful p-type doping¹⁻³ of the wide band gap II-VI material ZnSe, together with the discovery of quantum-well structures suitable for strong electron and hole confinement at and above room temperature,³ has enabled the recent realization of blue and blue green laser diodes.⁴⁻⁶ However, the difficulty in the fabrication of low-resistance ohmic contacts to the p-ZnSe layers is still an obstacle to be overcome before such devices can reach technological maturity. At present, the most commonly used method for contacting p-ZnSe involves the deposition of gold which forms a relatively low Schottky barrier contact; no suitable metal having a sufficiently high work function has been found.

We have recently reported that the same approach which has been found effective for the p-doping of Zn(S_{0.5}Se_{0.5}) by nitrogen, can be used to implement the most effective p-doping achieved for the molecular beam epitaxy (MBE) growth of ZnTe.⁷ Free-hole concentrations approaching 10^{19} cm^{-3} were easily obtained (the highest reported to date), while good crystalline quality, as revealed by x-ray rocking curves and photoluminescence measurements, was maintained. Our studies showed that gold forms a low-resistance contact to nitrogen-doped ZnTe epilayers, which naturally leads us to consider the use of heavily p-doped ZnTe as an intermediate layer for contacting p-ZnSe. Figure 1(a) shows the expected energy band lineup at a p-ZnTe/p-ZnSe interface, where it is seen that the valence band offset ($\sim 1 \text{ eV}$) between ZnTe and ZnSe forms a barrier to hole injection. A possible solution for removing the energy spike in the valence band is to introduce a Zn(S_{0.5}Te_{0.5}) layer having a graded band gap, as shown in Fig. 1(b). A similar contact scheme, but employing a graded (In,Ga)As region, was employed by Woodall *et al.*⁸ to form an ohmic contact between a metal-InAs junction and n-GaAs. It is interesting to note that the lattice constant mismatch is very similar between the binary compounds at the physical extremes in both the II-VI and III-V examples.

In this letter, we describe a quasiohmic contact to p-ZnSe which involves the injection of holes from heavily doped ZnTe into ZnSe via a pseudograded band gap region. A particular difficulty in realizing the band structure shown in Fig. 1(b) by MBE growth is controlling the Te fraction in the graded alloy region due to the competition between Te and Se species on the growth surface.⁹ To circumvent this problem, we have designed and grown a multilayer structure, consisting of 17 cells, each of 20 Å thickness. In each cell, both the thicknesses of ZnTe and ZnSe layers are varied, as shown in Fig. 1(c), to approximate a graded region. The first cell next to the p-ZnSe epilayer contained 18 Å of p-ZnSe and 2 Å of p-ZnTe; the next cell 17 Å of p-ZnSe and 3 Å of p-ZnTe, and so on.

The proposed contacting scheme was evaluated by means of I - V characterization, a transmission-line model

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FIG. 1. Schematic diagram of the energy band lineup at the (a) p-ZnTe/p-ZnSe interface and the (b) p-ZnTe/p-Zn(S_{0.5}Te_{0.5}) graded layer/p-ZnSe (c) Schematic drawing of the structure of the pseudograded Zn(S_{0.5}Te_{0.5}) contact layer

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FIG. 2 I - V characteristics of different contact schemes measured by an HP-4145 parameter analyzer at room temperature: (a) Au/ZnSe:Te-graded layer p -ZnSe, with the inset showing the I - V characteristic where the voltage range has a 50 mV scale maximum; (b) Au/ p -ZnSe; and (c) Au/ p -ZnTe/ p -ZnSe.

(TLM) measurement of specific contact resistance, and by employing the graded contact in laser diodes and LED devices. Techniques for the MBE growth and fabrication of laser diodes and LEDs have been previously reported;^{5,6,11,12} the details of ZnTe:N growth are described in a separate report.¹³

The contacts were first evaluated in a configuration designed for Hall measurements, as one objective was to obtain contacts suitable for the evaluation of the free-hole concentration in ZnSe:N epitaxial films. The nominally 4-mm-square samples had 0.8-mm-diameter contact pads in each of the four corners. Three different contacting schemes consisting of Au/ p -ZnSe, Au/ p -ZnTe/ p -ZnSe, and Au/ p -ZnTe/ p -Zn(Se,Te)/ p -ZnSe were evaluated. The p -ZnSe was a nominally 2- μ m-thick nitrogen-doped epilayer grown on an undoped semi-insulating GaAs substrate and had a hole concentration of $3.2 \times 10^{17} \text{ cm}^{-3}$. The I - V characteristics, measured between pairs of coplanar contact pads for the three contact configurations, are compared in Fig. 2. The hole concentration and the mobility of $14 \text{ cm}^2/\text{Vs}$ were easily determined from a conventional Hall measurement at room temperature when carried out using contacts formed by the graded layer. The ZnTe:N layer and the Zn(Se,Te) layer were etched down to the ZnSe:N layer everywhere except for the four Au contact areas in the configuration. As seen in Fig. 2(a), the graded contact appears to be perfectly ohmic, showing a straight line through the origin. The inset shows that the I - V characteristics maintain the same slope even at a few mV from the origin; the slope is maintained to the maximum test voltage of 100 V. Figure 2(b) shows the characteristics of the contact formed by gold deposited onto an as-grown ZnSe:N epilayer. The I - V characteristic corresponds to two back-to-back Schottky diodes; the observed turn on voltage is the reverse bias breakdown voltage. Increasing the doping level is expected to reduce the "turn-on" voltage of the contact. (Recently, the 3M group has observed that the turn-on voltage of the Au/ZnSe contact can be decreased by reducing the growth temperature of the up-

FIG. 3 I - V characteristics at 300 and 77 K from a structure having a Zn(Se,Te)-graded layer contact which is identical with Fig. 2(a), but with the contact size reduced to a 50 μ m dot.

permost region of the ZnSe.)¹⁴ Figure 2(c) shows the I - V characteristic when p -ZnTe is used to inject holes into the ZnSe epilayer in the absence of the graded region. For the structures of both Figs. 2(a) and 2(c), the ZnTe:N had a hole concentration of $5 \times 10^{18} \text{ cm}^{-3}$, as determined by Hall measurements. The deviation from ohmic behavior for the structure of Fig. 2(c) is attributed to the hole barrier arising from the valence band offset between the two semiconductors.

Figure 3 shows the I - V characteristics at 300 and 77 K from a structure identical to that of Fig. 2(a), but with the contact size reduced to 50 μ m in diameter and with current densities of up to 500 A/cm^2 ; some deviation from linearity is seen. We have observed that a Au/ p -ZnTe contact with an area roughly twice that of the 50 μ m dot also exhibits a deviation from linearity. Moreover, as it is shown in Fig. 1, the discontinuous nature of the grading plus the difference in doping levels between p -ZnSe and p -ZnTe layers are expected to cause a small effective energy barrier to holes, which contributes to the slight deviation from linearity. However, it is seen that the graded contact remains pseudo-ohmic at the lower temperature.

The specific contact resistance of the Au/ p -ZnTe-graded layer p -ZnSe contact was determined by a standard TLM measurement.¹⁵ An Ar ion beam mill was employed to isolate the TLM contact mesas down to the semi-insulating GaAs substrates. The Zn(Se,Te) graded layer between the Au electrodes ($50 \times 95 \mu\text{m}^2$) was also removed using the same milling technique. The specific contact resistance was determined to be in the range of 2 – $8 \times 10^{-3} \Omega \text{ cm}^2$. (A similar technique was applied to the Au/ p -ZnTe contacts, yielding a value of about $1 \times 10^{-3} \Omega \text{ cm}^2$.) The specific contact resistance of the graded contact was in the range (10^{-2} – $10^{-5} \Omega \text{ cm}^2$) usually considered acceptable for the contact area associated with LED and laser diode devices.¹⁶ One might well expect an improvement in performance as the growth is modified to more closely approach a continuous alloy grading and as doping levels are increased.

The Zn(S,Se)-based LED and laser device structures incorporated three (Zn,Cd)Se quantum wells (QW) of 75 Å thickness separated by 100-Å-thick Zn(S,Se). In these structures, we did not attempt the optimization of the op-

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FIG. 4. Light output characteristics of a (Zn,Cd)Se/Zn(S,Se) QW structure with a Zn(Se,Te) graded gap contact at 77 K. (a) Light output power of an LED as a function of current. The inset shows the I - V characteristics. (b) pulsed laser diode output power per uncoated facet as a function of current. The dark circles are some of the data points and the solid lines are to guide the eye.

tical and electronic confinement; rather, our emphasis was to show that the graded gap contact layer could withstand the high current densities of laser operation. Figure 4 shows the light output characteristics of a dc biased LED and a pulsed laser diode at 77 K. The LED forward conduction can be seen to onset at 3–4 V at 77 K in the inset of Fig. 4(a). Coincident with current turn-on, immediate visual evidence of LED action was observed: the output exceeds 100 μ W at 4 V. We concentrate here on the LED performance only at 77 K¹¹ since our earlier work has shown that, at this temperature, the quantum efficiency of the (Zn,Cd)Se QW emission is very high so that the recombination process is dominated by the radiative component.¹² This is an important point in the present context as we wish to show that the turn-on voltage for forward conduction results in a current which is usefully expended in supplying electron-hole pairs into the QWs for efficient light emission. Without such direct correlation, the possibility exists that an apparently low turn-on voltage could be due to a conduction mechanism which is influenced by defects in the multilayer ZnSe-based pn -junction devices. The efficiency of the LED was reduced by absorption in the top 2000-Å-thick ZnTe layer. For the laser diode with a graded contact, the threshold current density was approximately $J_{th} \approx 2$ kA/cm² (typically at a duty cycle of 1%). This threshold current is greater than we observed in previous devices⁵ and is the result of poor optical confinement in the particular experimental laser structures. In spite of the threshold current density, the forward bias voltage for lasing in these devices was reduced to 15–17 V (from ~30

V). Even with the nonoptimal design, these lasers have been operated at temperatures in excess of 200 K.

In conclusion, we have implemented ZnSe-based heterostructures where the electrical contact to p -type ZnSe has been substantially facilitated by the insertion of a p -Zn(Se,Te) pseudograded structure. The nearly ohmic characteristics of this contact scheme have been demonstrated together with their use in both blue/green light emitting diodes and diode lasers.

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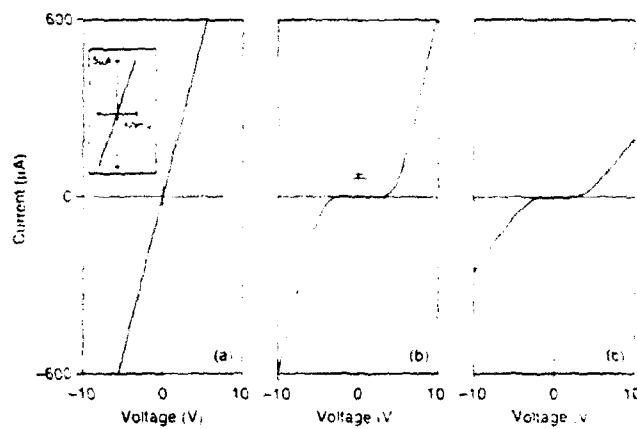
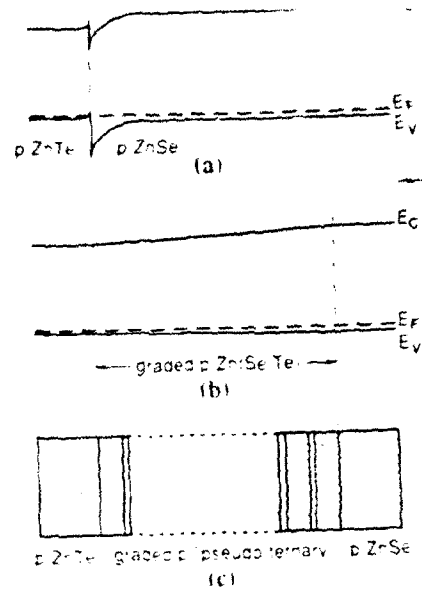
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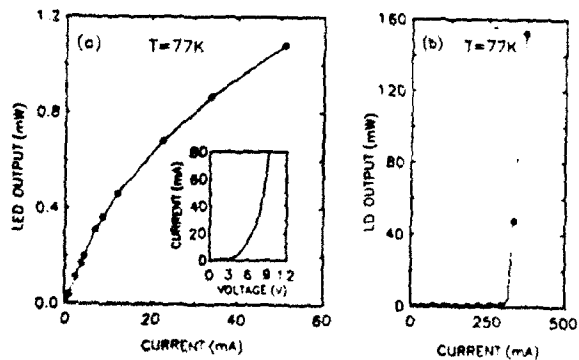
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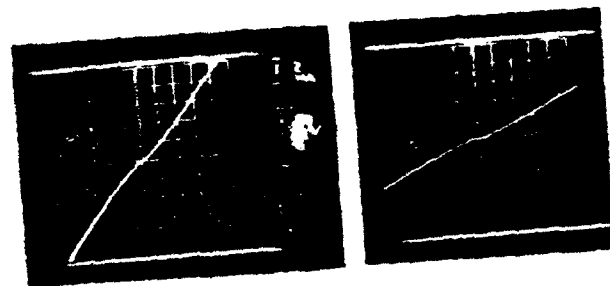
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(a) room temperature

(b) LN₂

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Blue and blue/green laser diodes and LED-based display devices

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We describe the performance of pn junction MQW diode lasers and LEDs which are grown on both p-type and n-type GaAs substrates. Efforts to minimize dislocations by lattice matching the II-VI region to the GaAs substrate in some cases resulted in obtaining dislocation densities below 10^5 cm^{-2} . We have obtained pulsed high power, high quantum efficiency laser emission up to room temperature conditions, and continuous operation at liquid nitrogen temperatures. Efficient LED and display devices operate in the blue (490–494 nm) at room temperature.

1. Introduction

Although early attempts to incorporate nitrogen by molecular beam epitaxy (MBE) were unsuccessful in obtaining p-type conductivity [1,2], recently Park et al. [3] and Ohkawa et al. [4], have obtained practical doping levels in MBE-grown ZnSe using a nitrogen plasma source made by Oxford Applied Research. Doping levels ranging from the mid 10^{17} cm^{-3} to low 10^{18} cm^{-3} , have led to the realization of pn junction light emitting devices operating in the blue and blue/green portion of the spectrum, such as the pulsed lasers reported in the summer of 1991 by 3M [5] and Brown and Purdue [6,7], and more recently by the group at Matsushita [8]. One of the first device configurations to exhibit lasing contains one or more (Zn,Cd)Se quantum wells imbedded within a ZnSe homojunction; the ZnSe is in turn bounded by Zn(S,Se) "cladding" regions. Lasing in such configurations have been reported by 3M, Brown and Purdue and, most recently, Matsushita. A difficulty with this structure is the inherent lattice parameter mismatch between the Zn(S,Se) and ZnSe layers; the result of the mis-

match is the existence of arrays of misfit dislocations at the two interfaces between these materials. The several device configurations described in this paper were designed to eliminate/minimize dislocations in the active II-VI regions.

2. MBE growth of device structures

Two different device configurations are described. The ZnSe-based structures have ZnSe/(Zn,Cd)Se multiple quantum wells (MQWs) sandwiched within a ZnSe p-n homojunction, while the Zn(S,Se)-based structures consist of Zn(S,Se)/(Zn,Cd)Se MQWs placed in a Zn(S,Se) p-n homojunction. The samples were grown in an MBE system equipped with separate growth chambers for the II-VI and III-V epilayer growth. For the ZnSe-based laser and LED structures, a 4 μm thick (In,Ga)As buffer layer was grown at a substrate temperature of 520°C. For the structures incorporating Zn(S,Se) layers, a 1.5 μm GaAs buffer layer was grown. The III-V buffer layers were transferred to the II-VI growth chamber under ultra high vacuum; the II-VI structures were nucleated at temperatures of 240–245°C. The Zn, Se, and Cd source materials were supplied by Osaka Asahi; the ZnS material

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was provided by Sumitomo Electric. Chlorine was the n-type dopant [9–11]; ZnCl_2 was used as the source of Cl. The flux ratios were all measured using a water cooled quartz crystal monitor placed at the substrate position.

3. ZnSe-based structures

The II–VI portion of the structures consisted of a p(or n)-ZnSe buffer layer ($0.1 \mu\text{m}$), up to 6 periods of a MQW structure consisting of 100 \AA ZnSe barriers and 60 \AA $\text{Zn}_{1-x}\text{Cd}_x\text{Se}$ ($x = 0.2$) wells, and an n(or p)-ZnSe top layer ($0.5 \mu\text{m}$).

Both (400) and (511) X-ray diffraction peaks were obtained, and used to evaluate the tetragonal distortion of the $(\text{In,Ga})\text{As}$ buffer layer. It was determined that the $\text{In}_x\text{Ga}_{1-x}\text{As}$ buffer layers were tetragonally distorted, since the lattice mismatch strain was only partially relaxed by misfit dislocations even though the thickness of the buffer layers was far greater than the predicted critical thickness. The c/a ratios were found to be approximately 1.001 [12]. The interfaces between the $\text{In}_x\text{Ga}_{1-x}\text{As}$ buffer layers with 4.3% In and the adjacent ZnSe were found to be nearly free from arrays of misfit dislocations. The lattice matching was obtained with a higher Th content than that expected from the lattice parameters of bulk crystals, a result consistent with the X-ray diffraction data.

TEM images revealed very low defect densities in the $(\text{In,Ga})\text{As}$ buffer layers. In cross-sectional images, segments of threading dislocations were found only in the regions close to the interface with GaAs. With plan-view imaging, no dislocations could be found in the upper parts of the buffer layers, suggesting that dislocation densities in those parts are in the range of 10^5 cm^{-2} or lower. Highly developed networks of misfit dislocations were observed at the $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ interfaces. Most of the misfit dislocations were found to have propagated towards the GaAs side rather than the $(\text{In,Ga})\text{As}$ side, implying a relative difficulty for dislocations to grow in $(\text{In,Ga})\text{As}$ crystals. The dislocation densities estimated from TEM images of the II–VI portion of the structures grown on $\text{In}_{0.043}\text{Ga}_{0.957}\text{As}$ buffer layers

were found to be in the lower range of 10^6 cm^{-2} . It is expected that the dislocation density could be reduced by further tuning of the In fraction.

It is significant that the confinement-enhanced exciton binding energy in these structures is approximately 40 meV, a value demonstrated to be sufficient to prevent dissociation from phonon interactions at temperatures above room temperature [13,14]. When operated as LEDs, we measure an external quantum efficiency of 10^{-3} at room temperature, despite the structures not having been optimized for reduction of total internal reflection losses.

The devices grown on p-III–V buffer layers are observed to exhibit considerable lateral conduction [15] and as a result require a mesa configuration for laser structures made from these samples. The laser devices consisted of $600 \mu\text{m}$ to 1 mm long cleaved resonator structures having $20\text{--}40 \mu\text{m}$ wide stripe electrodes at the top. Indium was evaporated as the contact for those structures having an n⁺-ZnSe top layer; gold was used to contact the devices having p-type top layers.

Laser emission was obtained under pulsed excitation, and for structures grown on both n- and p-type substrates.

At $T = 77 \text{ K}$, external differential quantum efficiency as high as 50% (both facets) has been obtained. The non-ohmic contacts contributed to typical turn-on voltages of $30\text{--}35 \text{ V}$ applied across the devices at laser threshold. Especially from the mesa devices, many transverse modes could be seen in the far field pattern. We also note that the laser emission occurs spectrally within the $n = 1$ HH exciton line and hence indicates that an excitonic component to laser action is present.

4. ZnSSe based structures

For the $\text{Zn}(\text{S,Se})$ based structures the II–VI portion consisted of a p-Zn(S,Se) buffer layer ($2 \mu\text{m}$), 6 periods of a MQW structure consisting of 100 \AA Zn(S,Se) barriers and 75 \AA $(\text{Zn,Cd})\text{Se}$ wells, and an n-Zn(S,Se) cap layer ($1.5 \mu\text{m}$). (Cited dimensions are nominal.)

Both cross-sectional and plan-view TEM images were taken. For some film growths no dislo-

$\text{In}_{0.043}\text{Ga}_{0.957}\text{As}$

cations could be found in cross-sectional samples; plan-view imaging was employed in order to evaluate the dislocation densities in those structures. The images show the Zn(S,Se)/GaAs interfaces to be free from misfit dislocations; some misfit (but no threading) dislocations are viewed at the MQW interface with the Zn(S,Se) buffer layer. The entire II-VI region below the MQW remains as a pseudomorphic layer. Fig. 1 shows a plan-view image of the Zn(S,Se)/GaAs interface. The dislocation density in the II-VI region was estimated at less than 10^5 cm^{-2} ; some samples probably have densities below 10^4 cm^{-2} .

The full widths at half maxima (FWHM) of {400} X-ray diffraction peaks obtained from the Zn(S,Se) layers with sulfur fractions between 7% and 8% ranged between 16 and 55 arc sec. These values are consistent with the low dislocation densities estimated from plan-view TEM imaging.

LED devices emitting in the blue (490 to 494 nm) at room temperature were prepared from $2 \times 2 \text{ mm}^2$ pieces, which were contacted by a 500 μm diameter indium dot. Blue LED emission could be readily obtained from the devices after a sufficient initial voltage was applied, typically 12–15 V, depending on the device. Due to the enhancement of the exciton binding energy by quantum well confinement in this wide gap II-VI

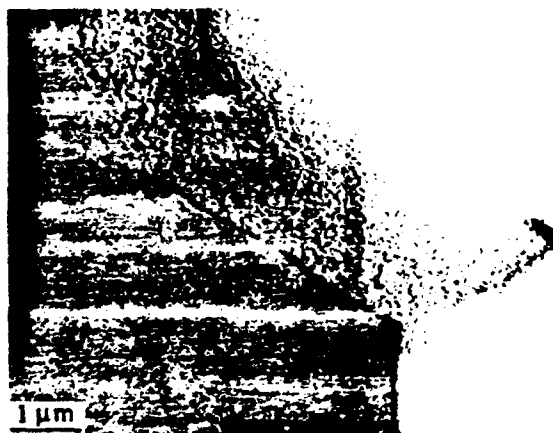


Fig. 1. Plan-view image of the interface between the GaAs epilayer and the Zn(S,Se) epilayer of a Zn(S,Se)-based laser structure. The lower Zn(S,Se) epilayer is seen to be pseudomorphic.

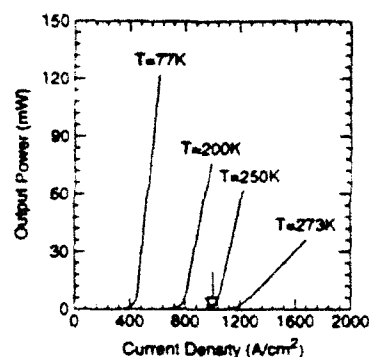


Fig. 2. Diode laser output power versus input current density for a mesa device with uncoated facets. The threshold current for this device at 77 K corresponded to a current of 160 mA. As room temperature is approached, heating, predominantly at the top contact, creates thermal problems. Except for the heating at close to room temperature, this device can be seen to exhibit a T_0 of about 180 K. The arrow indicates the threshold ($1 \text{ kA}/\text{cm}^2$) at 295 K for a mesa device with coated facets.

system, pairwise Coulomb correlations (excitons) can be maintained even at room temperature [14]. For a typical LED device, the voltage applied across the entire device corresponding to the highest light output power ($P = 120 \mu\text{W}$) was 20 V; most of this voltage is probably needed to overcome the built-in contact barriers.

Laser emission from both the mesa and the broad area devices was obtained under pulsed and CW excitation. Continuous wave operation at 77 K was obtained from a heat-sinked, Zn(S,Se)-based device fabricated on a p-GaAs substrate, with and without coated facets. Fig. 2 shows the pulsed diode laser output power versus input current density from 77 to 273 K for a Zn(S,Se) mesa device with uncoated facets. The threshold current density at $T = 77 \text{ K}$ was $400 \text{ A}/\text{cm}^2$, or 160 mA for the current of a typical device. At room temperature (at wavelengths of 490 to 494 nm), the threshold current density increased to $1500 \text{ A}/\text{cm}^2$ (corresponding to 600 mA actual current) for devices with uncoated facets. The arrow indicates the threshold ($1000 \text{ A}/\text{cm}^2$) at 295 K for a mesa device with coated facets. Various laser configurations have provided output powers in excess of 700 mW for pulsed operation at lower duty cycles. It is important to em-

phasize that lasing was obtained for structures grown on both n- and p-type substrates. Furthermore, it should be noted that the 3M group has also demonstrated pulsed lasing at 300 K, and CW lasing at cryogenic temperatures.

Acknowledgements

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